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Direct computation of structural parameter from diffraction pattern with multiple scattering

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Abstract : In this work, we report an attempt for determination of surface structural parameter directly from simulated single beam LEED intensity pattern from a class of possible structure We have made a detailed analysis of electron diffraction pattern with multiple scattering for one dimensional model of surface It has been shown that with some realistic assumptions, structural parameter can be directly determined from intensity pattern only for single relaxed overlayer structure.

Keywords : LEED, surface relaxation, electron-solid diffraction

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1. Introduction

Large scattering cross sections of atoms for low Energy electrons make LEED (Low Energy Electron Diffraction) a very powerful tool for surface structure determination [1]. But direct inversion of diffraction pattern to yield structure is difficult, as phase information is lost and strong multiple scattering introduces additional complexity. Therefore, surface structure are determined by method of searching among the set of plausible structures by comparing experimental and computed patterns [2]. In conventional LEED theory, a crystal is assumed to be consist of layers of ion cores parallel to the surface. Theoretical computation of LEED pattern from a given structure, firstly involves intraplanar multiple scattering, then layers are stacked one by one taking into account interplanar multiple scattering [3]. Here, we have performed our computation effectively for one dimensional model of surface, a chain of point scatterer. Intralayer multiple scattering is taken care of by energy dependent reflection and transmission coefficient of each scatterer. For one dimensional situation, a single beam intensity pattern is obtained whereas for three dimensional model, multiple beams one found.

In this paper, we examine the possibility of computation of structural parameter from diffraction pattern and successfully retrieve the relaxation parameter for some clean and adsorbed system with single relaxed overlayer from simulated diffraction pattern. To this end, loss of phase information is compensated by introduction of the constraint that the overall diffraction pattern must be compatible with complex reflection coefficient of ideally terminated bulk substrate, which can be computed from the known data of bulk lattice constant. We show that with this constraint, relaxation parameter can be directly obtained from the intensity pattern only for the case of single relaxed overlayer structure by inverting the equations of conventional LEED analysis. For two layer relaxed structure, intensity pattern as well as phase information is required for direct determination of structure. So reconstruction of phases of overall pattern is necessary for retrieval of parameters.

In Section 2, we present basic equations and line of computation and in Section 3, we present numerical results for some specific systems and discuss them. In final section, we conclude the paper with a summary of main points revealed in this work.

2. Theory

Reflection coefficient R_L of a model of surface with one relaxed layer [4] at a distance d_{12} over the substrate having complex reflection coefficient R_S is

$$R_L = r + \frac{R_S t^2 e^{ik2d_{12}}}{1 - rR_S e^{ik2d_{12}}}, \tag{1}$$

k is the wave vector of incident electron beam. Interaction of incident electrons with relaxed scatterer can be summarised in terms of its complex reflection and transmission coefficients r and t respectively and these depend on chemical composition and two dimensional geometry of the layer. R_S can be computed by layer doubling method [3], converging the incident electron beam within a stack of identical layers to the semi-infinite crystal limit, *i.e.*, by iterating eq. (1) using the bulk lattice constant a for d_{12} and R_S for R_L, taking the complex reflection coefficient and transmission coefficient of the bulk specimen r_b and t_b respectively. From eq. (1), it is clear that the relaxation parameter d_{12} for a single relaxed structure can be determined easily if both the phases and amplitudes of R_S and R_L are known, when chemical composition and layer geometry of bulk and relaxed layer are given. But as phases of R_L are lost in experiment, we eliminate the phases of R_L from eq. (1) and obtain

$$\cos\left(2kd_{12} + \phi + \theta\right) = \frac{|R_L|^2\left(1+|R_S|^2|r|^2\right) - |r|^2\left(1+|R_S|^2|r|^2 \left\| \frac{t^2}{r^2} - 1 \right|^2\right)}{2|R_S||r|A}, \tag{2}$$

where A is computed from the equations

$$A\sin\theta = |t^2 - r^2|\sin\varsigma, \qquad A\cos\theta = |R_L|^2 + |t^2 - r^2|\cos\varsigma,$$

when $\phi = \arg(R_S) + \arg(r)$, $\varsigma = \arg\left(t^2/r^2 - 1\right)$.

$|R_L|$ is known from diffraction intensities and R_S, ϕ and θ can be computed from a, the bulk lattice constant and $|R_L|$. Therefore, the relaxation parameter d_{12} can be determined. For a fixed value of right hand side of eq (2), two cosine angles (say, α and $2\pi - \alpha$) can be obtained. With the physical restriction that d_{12} is close to the bulk lattice constant a, four possible values can be obtained at each energy grid point. Of these four possible distances, two are contractive ($d_{12} < a$) and other two are expansive ($d_{12} > a$) Two values of d_{12} are computed from the angle α and the other two are from $(2\pi - \alpha)$. At each energy, we get a set of four different d_{12}s, the unique value of these sets gives the actual value of d_{12}. So for surfaces with one relaxed overlayer, either clean or adsorbed, relaxation parameter can be determined from intensity pattern only so that it must reproduce substrate reflection coefficient properly in full range of energy

For surfaces with two relaxed overlayer, an intermediate layer comes between to layer and bulk periodic substrate. Let R_{S1} be the complex reflection coefficient of that intermediate layer with regular substrate underlying. This R_{S1} is not known, hence d_{12} can not be computed from the intensity pattern alone. However, if the phases along with intensity is available, then $|R_{S1}|$ can be determined from the equation

$$|R_{S1}| = \frac{R_L - r}{t^2 - r^2 + rR_L} \tag{3}$$

Once the $|R_{S1}|$ is known, d_{23}, the distance between second and third overlayer can be obtained by the same way as it was determined for single overlayer structure. Then phases of R_{S1} can be computed. Therefore, in the next step, the relaxation parameter d_{12} can also be obtained.

Recently, the phase information has been recorded experimentally in holographic LEED methods for some special system [5], but the real difficulty is that phases are not available in most of the experiments. On the other hand, phases must be specific so that the reflection coefficient of the bulk can be matched with two real distances d_{12} and d_{23} in successive application of eq. (1). So reconstruction of phase information of overall reflection coefficient R_L is worthwhile to compute the relaxation parameters. In the next section, we present the results and discuss their importance and implication.

3. Results and discussion

Our technique successfully retrieved the relaxation parameter from simulated experimental data of clean surfaces Al (111), Rh (111), Cu (111), Pd (100) and oxygen absorbed system of Al (111), Rh (111). Energy dependent reflection and transmission coefficients of a plane for each type of species are computed from standard dynamical LEED code [4] for –(00) beam of incident electron. Standard energy dependent atomic phase shifts, bulk interplanar distance and two dimensional lattice vectors of a plane, are taken as inputs for this computation. We have considered intensity pattern for different single overlayer structure with various values of d_{12}, both contractive and expansive relaxation with typical variation of bulk interplanar distance and these simulated intensities are taken as pseudo

experimental data This simulated intensity pattern, bulk lattice constant and energy dependent r and t are taken as inputs for this direct computation. In Table 1, we have shown some typical values of parameters of surfaces for simulating diffraction pattern and the retrieved parameters.

Table 1. Typical values of parameters of surfaces for simulating intensity patterns and the retrieved parameter values

Surfaces	Bulk interplanar distance (a) (Å)	Simulated value of d_{12} (Å)	Retrieved value (Å) of d_{12} (Å)
Al (111)	2.338	2.216	2.216
Cu (111)	2.087	2 034	2 033
Rh (111)	2.192	2.246	2.246
Pd (100)	1 945	2 003	2 002
Al (111) + (1X1)O overlayer	2.338	1 462	1.462
Rh (111) + (2X2)O overlayer	2.192	1.238	1.237

As we have pointed out for surface with two relaxed overlayer, exact structural parameters can be determined if the correct phase information of the top layer can be somehow reconstructed. It seems that understanding of phase variation with energy and its relation with R_S is essential for making a progress in this direction. We have studied previously [6] the intensity *vs* energy curve (I–V curve) and parametric plots (real *vs* imaginary plot) of R_L and R_S of some one dimensional structures with several relaxed overlayers taking energy independent typical values of $r = 0.1$ and $t = 0.9 + 0.4i$ [3] for simplicity. Study of those intensity curve reveals that for simple relaxed overlayer structure, position of Bragg peak do not change appreciably as that of bulk terminated periodic substrate, but extra features appear in the region between two Bragg peaks. Therefore, near Bragg peaks patterns for multilayer relaxed structure can be considered as an effective single relaxed overlayer structure with a modified R_S. So an iteration may be employed for reconstructing the phases of R_L with the help of eq. (2) and probability of convergence of such iteration will be maximum at the energy points near Bragg peaks. Further investigation is required to establish the nature of this iteration.

4. Conclusion

We have shown here that relaxation parameter for a single relaxed overlayer structure can be determined directly from diffraction intensity pattern only even in presence of multiple scattering. Additional constraint that the diffraction intensity should be compatible with complex reflection coefficient of the bulk periodic substrate, compensates the loss of phase information. Though this one dimensional model appears to be simple in current framework of LEED theory, but this work shows a clear approach of direct determination of structure without trial and search procedure. This work should be extended for general multilayer

relaxed structure with three dimensional multibeam effect for direct inversion of real experimental LEED pattern.

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